DYE LASER AS A SIX-LEVEL SYSTEM

By

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Stationary solutions of kinetics equations of dye laser based on a six-level model of dye molecule are analysed. Such a model is the simplest one to account for the participation of triplet states in generating a laser. The course of triplet losses versus pumping parameter can undergo a jump at threshold which leads to the jump of photon number in the cavity. The improvement of the laser performance of dye lasers by means of an additional triplet-triplet pumping is proposed.

The mathematical description of a commonly used dye laser system is based on an approximation in which the population of higher triplet states is neglected. Usually this approximation holds when the molecular system is not exposed to strong electromagnetic fields. When the molecular system is an active medium of a dye laser this approximation is questionable because of a high number of photons in the cavity. In this case, even when the depopulation rates of higher triplets are of the order of 10^{11}—$10^{12}\,s^{-1}$, the populations of these states cannot be neglected. The simplest model for a dye molecule which accounts for the laser action in a dye laser should be a six-level model, given in Fig. 1. Singlet electronic and vibronic states are denoted by E_1, E_2, E_3, E_4, triplet states by E_5 and E_6. The action of such a laser system was analysed in [1—3]. The quantum mechanical theory of dye lasers with a six-level model of the

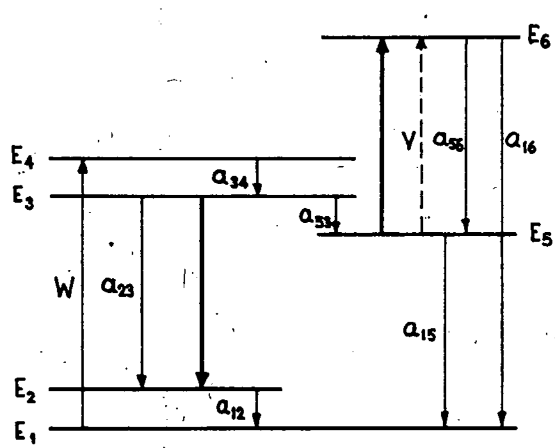

Fig. 1. Energy levels diagram of a dye molecule.
W — pumping parameter; *V* — additional triplettriplet pumping parameter

active medium in adiabatic approximation leads to the system of kinetic equations describing the time evolution of photon number in the cavity and that of the populations of energy levels of the dye molecule. The stationary solution of these equations leads to the dependence of photon number n on pumping parameter W in the form

$$W = \frac{(1+an)(1+bn)}{-An^2+Bn+C}, \tag{1}$$

where

$$a = \frac{b_1}{a_{23}+a_{53}}\left(1+\frac{a_{53}}{a_{12}}\right), \quad b = \frac{b_2}{a_{16}+a_{56}}\left(1+\frac{a_{16}}{a_{15}}\right),$$

$$A = \frac{ab}{a_{12}+a_{53}}\left(\frac{2a_{53}}{a_{15}+a_{16}}+\frac{a_{12}+a_{53}}{a_{34}}+2\right),$$

$$B = \frac{ab(a_{16}+a_{56})}{2\varkappa(a_{12}+a_{53})}\left(\frac{a_{12}-a_{23}}{a_{16}+a_{56}}-\frac{a_{53}}{a_{15}+a_{16}}\right)-$$

$$-\frac{a}{a_{12}+a_{53}}\left(\frac{a_{53}}{a_{15}}+\frac{a_{12}+a_{53}}{a_{34}}+2\right)-$$

$$-\frac{b}{a_{23}+a_{53}}\left(\frac{a_{23}}{a_{12}}+\frac{a_{23}+a_{53}}{a_{34}}+\frac{2a_{53}}{a_{15}+a_{16}}+1\right),$$

$$C = \frac{a}{2\varkappa}\cdot\frac{a_{12}-a_{23}}{a_{12}+a_{53}}-\frac{b}{2\varkappa}\cdot\frac{a_{53}(a_{16}+a_{56})}{(a_{23}+a_{53})(a_{15}+a_{16})}-$$

$$-\frac{1}{a_{23}+a_{53}}\left(\frac{a_{23}}{a_{12}}+\frac{a_{23}+a_{53}}{a_{34}}+\frac{a_{53}}{a_{15}}+1\right). \tag{2}$$

Quantities a_{ij} in (2) are transition rates indicated in Fig. 1, b_1 and b_2 are Einstein coefficients for $2 \leftrightarrow 3$ and $5 \leftrightarrow 6$ transitions, respectively and $2\varkappa$ describes cavity losses. It is to be noted that quantities a, b, A, B, C depend only on molecular and cavity parameters. The transition rate a_{16} is introduced to account for processes of depopulation of higher triplets of other than $6 \to 5 \to 1$ transition.

It is shown [1—3] that there exist two main kinds of n–W characteristics: one (Fig. 2A) in which the photon number changes monotonically and continuosly from value zero, typical also for three and four level systems, and the second (Fig. 2B) characteristic typical only for a six-level system, where the photon number undergoes a jump at the threshold. Depending on the choice of parameters describing a dye laser system one of the n–W characteristic is to be expected. It was shown, based on the stochastic theory [2], that the n–W characteristic given in Fig. 2B could be thought of as a phase transition analogy of the first order in a laser system.

These two types of solutions can be interpreted by means of triplet losses in the active medium. The definition of triplet losses given in [3] reads

$$T(n, W) = b_2(p_5-p_6), \tag{3}$$

where p_5 and p_6 are populations of triplet states E_5 and E_6, respectively. In Fig. 3 triplet losses are presented for both types of solutions. As the populations of triplet sates depend not only on pumping parameters but also on the number of photons in the cavity, triplet losses depend on generation conditions. The n–W characteristic with

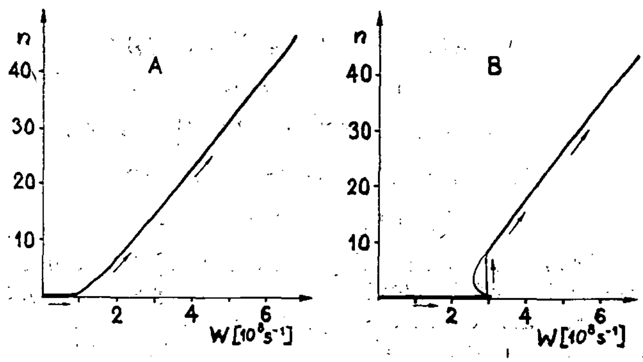

Fig. 2. Dependence of photon number n on pumping parameter W for the dye laser system characterized by $a_{12}=a_{34}=a_{56}=10^{11}\,s^{-1}$, $a_{23}=2\cdot10^8\,s^{-1}$, $a_{53}=10^8\,s^{-1}$, $a_{15}=0.6\cdot10^7\,s^{-1}$, $a_{16}=10^9\,s^{-1}$, $2\varkappa=10^7\,s^{-1}$, $b_2=4\cdot10^7\,s^{-1}$, $b_1=1.2\cdot10^7\,s^{-1}$ (A), and $b_1=10^7\,s^{-1}$ (B)

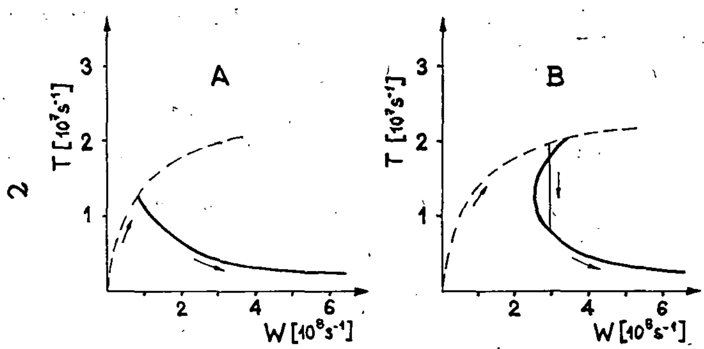

Fig. 3. Dependence of triplet losses T *vs.* pumping parameter W for the system specified in Fig. 2. Dashed lines are indicating the triplet losses for trivial solutions ($n=0$)

a jump at the threshold is to be expected only in such a case when triplet losses undergo a jump (Fig. 3B).

There exist a number of dye-laser systems which do not lase by flash-lamp pumping, because of the triplet losses. These losses can be reduced by means of an external photon beam which causes a depopulation of the lower triplet state. Kinetic equations of a six-level system with additional triplet-triplet pumping have the following form

$$
\begin{aligned}
\dot{n} &= -2\varkappa n + b_1(p_3 - p_2)n - b_2(p_5 - p_6)n, \\
\dot{p}_1 &= a_{12}p_2 + a_{15}p_5 + a_{16}p_6 - Wp_1, \\
\dot{p}_2 &= a_{23}p_3 - a_{12}p_2 + b_1(p_3 - p_2)n, \\
\dot{p}_3 &= a_{34}p_4 - (a_{53} + a_{23})p_3 - b_1(p_3 - p_2)n, \\
\dot{p}_4 &= Wp_1 - a_{34}p_4, \\
\dot{p}_5 &= a_{53}p_3 + a_{56}p_6 + b_2(p_6 - p_5)n - (V + a_{15})p_5, \\
\dot{p}_6 &= Vp_5 - b_2(p_6 - p_5)n - (a_{56} + a_{16})p_6, \\
& p_1 + p_2 + p_3 + p_4 + p_5 + p_6 = 1.
\end{aligned}
\tag{4}
$$

Quantity V describes the $5 \to 6$ transition rate caused by the additional T—T pumping. Stationary solutions of Eqs. (4) is similar to those given by (1), but parameters A, B, C and b depend also on V. Usually, the additional T—T pumping improves laser performance of dye lasers, specially in systems with high triplet losses. It is to be pointed out, that the additional T—T pumping may not occur exactly at the generation frequency. This fact can be easily incorporated into the present theory by introducing an additional triplet state. It seems highly probable that the additional T—T pumping takes place in the usual broadband flash-lamp pumping of dye lasers.

The influence of additional T—T pumping on n–W characteristics is presented in Fig. 4. It is to be noted that the choice of parameters was such that with $V=0$ no laser action was possible. The influence of additional T—T pumping on the laser performance of dye lasers was observed experimentaly. Results of these investigations will be published elsewhere.

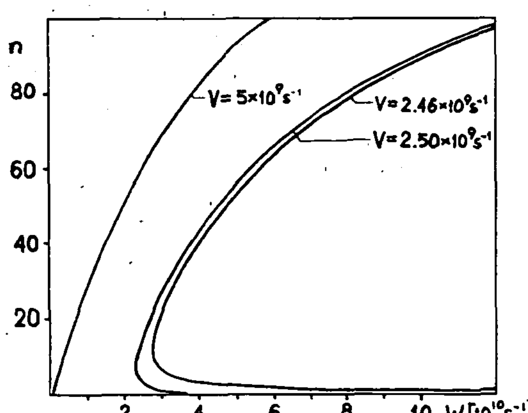

Fig. 4. The influence of additional triplet-triplet pump V on $n-W$ characteristics of stationary solutions of dye lasers. The molecular and cavity parameters have the following values: $a_{12}=a_{34}=a_{56}=10^{11}\,s^{-1}$, $a_{23}=2\cdot10^{8}\,s^{-1}$, $a_{15}=0.6\cdot10^{7}\,s^{-1}$, $a_{16}=10^{9}\,s^{-1}$, $b_1=8\cdot10^{7}\,s^{-1}$, $b_2=4\cdot10^{7}\,s^{-1}$, $2\varkappa=5\cdot10^{7}\,s^{-1}$.

References

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ЛАЗЕР НА КРАСИТЕЛЕ КАК ШЕСТИУРОВНЕВАЯ СИСТЕМА

А. Бачински и А. Косановски

В работе анализируются стационарные решения кинетического уравнения лазера на красителе, основанные на шестиуровневой модели молекулы красителя. Такая модель является наиболее простой, учитывающей участие триплетных состояний в генерации. Показано, что вблизи порога ход триплетных потерь в зависимости от параметров накачки может испытывать скачок, который приводит к резкому увеличению числа фотонов в резонаторе. Предложен метод улучшения характеристик лазера на красителе путем допольнительной триплет-триплетной накачки.